AD 746148

PAPER P-847

ELECTRICAL HIGH-PRESSURE MOLECULAR GAS LASERS

E. J. Seppi

May 1972

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UNCLASSIFIED

Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1 ORIGINATING ACTIVITY (Corporate author) INSTITUTE FOR DEFENSE ANALYSES 400 Army-Navy Drive Arlington, Virginia 22202	2a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED 2b GROUP --

3 REPORT TITLE

Electrical High-Pressure Molecular Gas Lasers

4 DESCRIPTIVE NOTES (Type of report and inclusive dates)

Paper P-847, May 1972

5 AUTHOR(S) (First name, middle initial, last name)

E.J. Seppi

6 REPORT DATE May 1972	7a. TOTAL NO OF PAGES 42	7b. NO OF REFS 84
8a CONTRACT OR GRANT NO DAHC15 67 C 0011 b. PROJECT NO DARPA Assignment 5 c d	9a. ORIGINATOR'S REPORT NUMBER(S) P-847 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) None	

10 DISTRIBUTION STATEMENT

Approved for public release; distribution unlimited.

11 SUPPLEMENTARY NOTES NA	12 SPONSORING MILITARY ACTIVITY DEFENSE ADVANCED RESEARCH PROJECTS AGENCY Arlington, Virginia 22209

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DD FORM 1 NOV 68 1473

UNCLASSIFIED

Security Classification

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Laser						
Molecular laser						
Gas laser						
Electrical laser						
High-pressure laser						
Laser design						

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Contract DAHC15 67 C 0011
DARPA Assignment 5

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ACKNOWLEDGMENT

The author wishes to thank Dr. John Asmus for suggesting the study of electrical high-pressure molecular lasers and also for reading and commenting on the final manuscript. Appreciation and acknowledgment are also given to Dr. Robert Fox for making available the summer position at IDA during which this study was prepared.

ABSTRACT

The design and operation of high-power, electrically excited gas lasers are discussed with particular attention given to molecular-type lasers operating at high pressure. A survey is presented of gases in which laser action has been initiated by electric discharge at high gas pressure. The basic processes for operation of the carbon-dioxide and carbon-monoxide lasers are described. The design criteria and the potential for high power and high brightness from a carbon-dioxide laser are discussed. The engineering requirements for a pumping arrangement which uses an electron beam for pre-ionization to obtain uniform electric pumping are reviewed.

CONTENTS

I. INTRODUCTION

Until recently gas lasers as a class have been much less powerful and less efficient than solid state lasers. However, several developments have resulted in substantial performance advances in the construction of high-power, high-brightness gas lasers[1,2,3,4]. Powerful pulsed gas lasers can now be designed which are competitive with solid state units. Existing gas lasers can be classified into three broad classifications, according to the principal active species, as ionic atom, neutral atom, and molecular. Typically ion lasers operate in the wavelength region spanning the visible region; neutral atom lasers operate in the visible and near infrared; and molecular lasers operate in the far infrared.

Known neutral atom (for example the helium-neon laser) and ionic atom (for example the argon laser) devices have relatively low efficiencies for converting electrical input energy into laser light. Efficiencies of the order of 0.03 percent are typically observed. On the other hand for molecular lasers using CO_2 or CO efficiencies of the order of 30 percent and above have been achieved. This is a three order of magnitude increase in efficiency over that of previous gas systems and approximately one order of magnitude higher than that achieved in optically pumped, solid state lasers.

Typical gas pressures in longitudinal electric discharge lasers are less than a few torr for atomic and ionic lasers and less than a few deca-torr for molecular lasers. The operation of gas lasers at higher pressures near or above atmospheric pressure results in a further increase in the specific power which can be obtained from a gas device. Gas lasers using CO_2 with various combinations of other gases operating near atmospheric pressures have been studied using both high

voltage techniques in the standard longitudinal gas discharge configuration[5] (Hill) and the transverse electric field technique at atmospheric pressures (TEA) which was developed by Beaulieu[6] and studied by numerous authors[1,7-15]. This latter technique greatly simplifies the equipment required to study the high-pressure gas laser. Using the technique CO_2 laser pulses of energy comparable to solid state devices have been obtained. Pressures of approximately one atmosphere which are presently being used are not the upper limit. If suitable means of pumping can be developed laser operation of gas pressures of 10 to 100 atmosphere may be possible. This combined with efficiences in the 10-40 percent range will result in pulsed gas lasers which compete strongly with solid state lasers for the production of high peak power pulses.

The typical TEA arrangement is not suitable for production of a uniformly excited medium over a large cross-sectional area necessary to reliably achieve laser power of high optical quality. R. Dumanchin[1,16] has reported achieving 130j output in a 2 microsec pulse from an electrically pulsed CO_2 laser. The laser used 20 liters of gas at atmospheric pressure and is uniformly electrically pumped by using pre-ionization techniques. Jack Dougherty* and his coworkers are actively pursuing the use of pre-ionization techniques in which a pulsed high-energy electron beam is injected into the active region of the laser gas to produce ionization electrons. These electrons are accelerated in a DC electric field to attain inversion in a CO_2-N_2-H_e gas mixture. Preliminary tests indicate that uniform pumping over large volumes can be achieved with this technique.

A number of experimenters have used gas systems which flow gas through the cavities[17]. Using this technique some interesting methods have been employed for obtaining population inversions which include rapid fluid mixing, gas dynamic expansion[18] and chemical reaction pumped gas lasers. Typical arrangements for a flow system are shown in Figs. 1a and 1b.

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*Private communication.

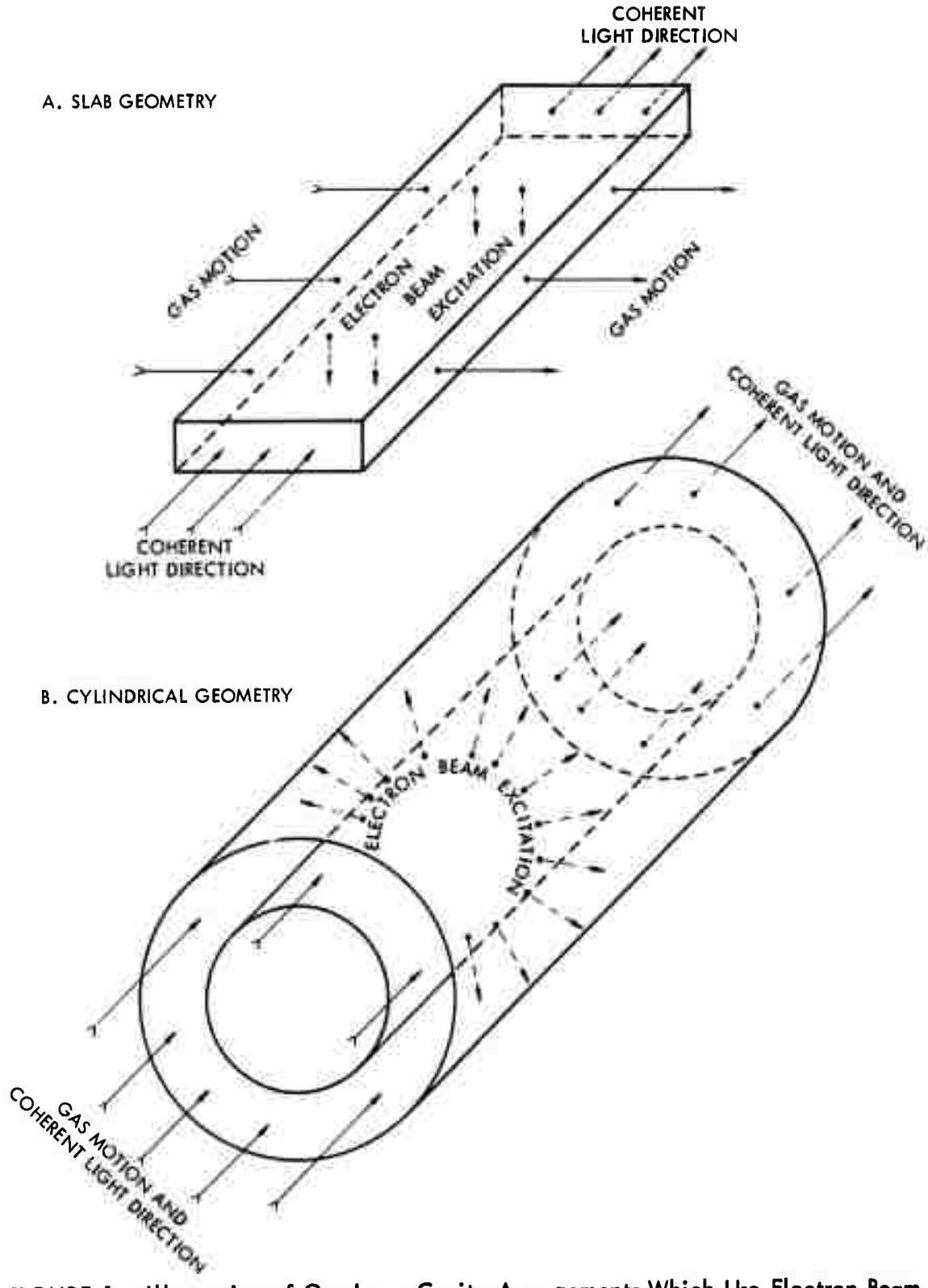

FIGURE 1. Illustration of Gas Laser Cavity Arrangements Which Use Electron Beam Excitation and Gas Flow. (Schematics A and B show, respectively, a slab and cylindrical geometry.)

The important features of the gas flow system are:

1. If desired it allows for separation of the excitation process from the relaxation processes. This allows for various pumping schemes as previously indicated.
2. Perhaps more important, the high-speed flow provides a means to exhaust unusable energy from the active cavity and, thereby, permits the minimization of the undesirable effects on gain and laser operation which result from heating of the material in the laser cavity. In non-flowing lasers these effects are greatly influenced and controlled by cavity wall geometry and surface. In the flow system the effect of the wall geometry is essentially eliminated. Flowing gas systems can thus be designed to permit higher CW power or higher repetition rate for pulsed systems. Also, these systems provide favorable scaling characteristic which allow for the increase in the volume of gas used in the laser.

II. SURVEY OF LASER ACTION INITIATED BY ELECTRIC DISCHARGE IN VARIOUS GASES AT HIGH-PRESSURE

As previously mentioned, CO_2 is the most common gas which has been used as the laser medium in high-power high-pressure lasers. However, a wide variety of gases have produced laser action at near atmospheric pressure. Although considerable effort is presently being expended on the development of the CO_2 system for high power and high brightness there is no reason to not expect additional important discoveries comparable to that of the CO_2 laser. Other gases which have been observed to lase using pulsed transverse excitation are given in Table 1. Transitions in the region from 0.8 to 28μ have been observed. Some gases have been observed to operate at near atmospheric pressure and promise to give high-power output. The last column of the table gives roughly the relative power output of the various transitions observed in a standard transverse excitation cavity. In some cases a peak-power output is listed. Although the authors of the studies of these gases, in general, have attempted to optimize the output they caution that the experimental arrangement did not allow complete optimization of all parameters. Therefore, the last column of the table may contain erroneous information. Note that considerable output was obtained from HF and N_2O. Recent work on CO has indicated that high efficiencies and high power are possible from this gas. The potential of efficiencies higher (theoretically over 80 percent) than those observed in CO_2 and the possibility of high-power laser beams in the 5.2-5.7μ region make CO an attractive candidate for further study.

Intense super radiant emission has been observed in the infrared transitions of HF and DF[21,23]. Most of the work with HF and DF has studied the laser emission in the 2.7 and 3.8μ range, respectively, as a result of chemical reaction excitation as opposed to electrical

TABLE 1. GASES DEMONSTRATING LASER ACTION AT HIGH PRESSURE WITH TRANSVERSE EXCITATION[19-22]

Laser Gas	Typical Total Pressure X+He	Added Gas X(% total)	Wavelength	Relative Output Power
		Atomic Transitions		
He	65 Torr	NH_3 (18%)	2.06 μm	Weak
C	250	CO or CH_4	1.45	Medium
O	75	O_2 (1%)	0.845	Weak
Ne			1.15	Weak
Ne			1.20	Weak
Ne	30	Ne (30%)	1.25	Weak
Ne			1.52	Weak
Ne	120		3.39	Strong
Cl	250	NOCl (1%)	1.59	Medium
Ar	100		1.27	Weak
Ar	760		1.79	Strong (0.4 kw)
Ar	100	Ar (30%)	2.21	Strong
Ar			2.39	Strong
Ar			5.80	Medium
Ar			7.29	Medium
Kr	760	Kr (7%)	2.52	Strong
Kr			3.07	Strong
Xe	250	Xe (5%)	2.03	Strong (0.4 kw)
Xe			3.51	Medium (0.1 kw)
Xe			3.65	Strong (0.5 kw)
		Molecular Transitions		
HF	100	SF_6 (3%)	12.67	Weak
HF		$+H_2$ (<1%)	13.19	Medium
HF			13.78	Weak
HF			14.44	Strong
HF			15.17	Weak
HF			2.78-3.00	Weak
HF	400		2.76-3.09	Very Strong (40 kw)
HF	100		2.82-3.05	Weak
DF	10-300	SF_6:D_2 ~ 10:1	3.8	Strong
CO	200	CO (30%)	5.21-5.72 (44 lines)	Strong
N_2	250	N_2 (23%)	1.04-1.05 (7 lines)	Strong (0.2 kw)
N_2			1.23-1.25 (5 lines)	Medium
N_2			3.64-3.65 (3 lines)	Medium
H_2O	45	H_2O (>95%)	7.60	Weak
			7.71	Weak
			9.39	Weak
			9.47	Weak
			9.57	Weak
			27.97	Strong
CO_2	450	CO_2 (3%)	9.28-9.31	Medium
			9.54-9.57	Medium
		CO_2 (25%)	10.26-10.30	Medium
	760		10.52-10.63	Very Strong (96 kw)
N_2O	300	N_2O (10%)	10.48-10.55	Medium
			10.77-10.86	Very Strong (6.0 kw)
SiO_2	2	SiO_2 (50%)	140-215	Weak

excitation. In these studies a u.v. source or electronic discharge is used to initiate the chemical reaction. Several reactions have been studied[24-27]; some gas combinations which have been experimented with include: NF_3 or N_2F_4 mixed with H_2, CH_4, C_2H_6, HCl, HBr or natural gas; CF_4 or CCl_2F_2 mixed with H_2, D_2 or CH_4; and SF_6 mixed with H_2 or D_2. In some cases He was added to the mixture to obtain more intense radiation.

The chemical reactions which take place in these electric discharge initiated chemical lasers are in general a complicated system. For example, in the NF_3-HX system where X = H, CH_3, C_2H_5, Cl or Br, the following reactions take place:

$$NF_3+\bar{e} \rightarrow NF_2+F+\bar{e} \qquad (1)$$

$$\rightarrow NF+F_2+\bar{e} \qquad (2)$$

$$F+HX \rightarrow HF+X \quad \Delta H \cong -48\text{Kcal/mole} \qquad (3)$$

$$H+NF_3 \rightarrow HF+NF_2 \quad \Delta H \cong -77\text{Kcal/mole} \qquad (4)$$

$$H+NF_3 \rightarrow HF+NF \quad \Delta H \cong -65\text{Kcal/mole} \qquad (5)$$

$$2NF \rightarrow N_2+2F \quad \Delta H \approx -82\text{Kcal/mole} \qquad (6)$$

These reactions can result in HF excited into vibrational modes which yield inversion for lasing. However, the various mechanisms involved and the dependence of the various reaction rates are not well understood and as a result optimization of laser output is difficult. It appears that poor efficiency in population of the upper laser levels and rapid depopulation of these levels by routes other than stimulated emission result in low overall efficiences for generating optical laser powers. In fact, completely neglecting the chemical reaction energy the highest "efficiency" for energy output to electrical energy input that has been observed in this type of laser is less than 1 percent. Using a mixture of SF_6 and H_2 with partial pressures of 300 and 10 torr, respectively, a peak power of 32 kw per meter of discharge has been obtained[21]. The high-energy content which can be liberated in chemical reaction makes the chemical laser an attractive device. However, the necessity for chemical reaction in or

near the active region of a laser can only serve to complicate the overall operation and optimization of the laser by adding another function in a region already complicated by the requirements of the laser for inversion and optical homogeneity.

III. DESCRIPTION OF BASIC PROCESSES FOR OPERATION OF MOLECULAR LASERS

Two basic schemes for obtaining the population inversion necessary for laser operation in molecular lasers are illustrated by the CO_2 and the CO lasers. In typical electrical CO_2 laser operation a true inversion of the total population of the various vibrational modes of the ground electronic state can be achieved. Since CO is diatomic only one vibrational mode is available. In this case typical CO laser operation depends on inversion in the rotational-vibrational levels and does not necessarily require inversion of the total population of the individual vibrational states. Below we briefly describe the operation of the CO_2 and CO processes for molecular laser operation. Other more complex schemes are also possible; for example, processes involving the numerous vibrational modes of polyatomic molecules which have nonlinear ground states are possible but will not be discussed.

A. MECHANISM OF OPERATION OF THE CO_2 LASER

The decay and excitation scheme for the operation of the CO_2 laser is r sonably well understood and has been described by a number of authors[28,29]. However, details of the various mechanisms involved can vary considerably from one laser configuration to another making inter-comparisons difficult without detailed analysis of the operating conditions. The CO_2 molecule in its ground electronic state can be described as a linear molecule and has three vibrational modes. This allows laser operation through inversion and stimulated decay between the rotational-vibrational levels in different vibrational modes[30-32]. This type of operation is possible only in polyatomic gases containing three or more atoms, and can be considered as a general example of possible laser mechanism for this case. Figure 2a schematically shows the vibrational modes and

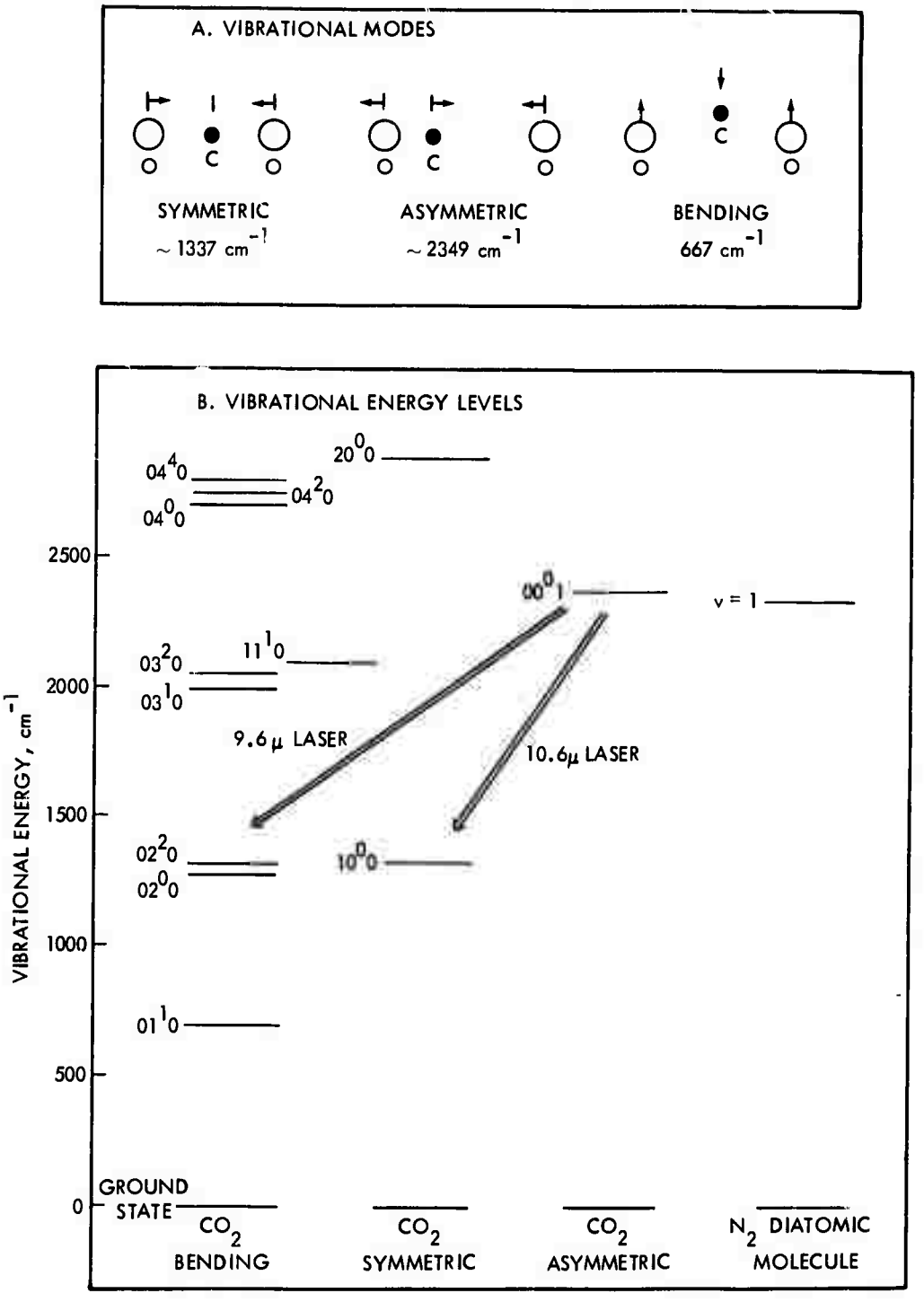

FIGURE 2. Vibrational Modes of CO_2 and N_2 Molecules Important in Gas Laser Operation. (Rotational states and many higher levels of excitation of the molecules are not shown.)

characteristic frequencies of the CO_2 molecule for its lowest electronic state. Figure 2b shows the lower vibrational energy levels of CO_2. Each vibrational level is further split into a collection of closely spaced rotational levels. Also shown on the figures are N_2 vibrational levels which are significant in the laser action of a N_2-CO_2 system. When energetic electrons collide with the CO_2 molecule it is excited into higher electronic and vibrational-rotational levels. In the process a population inversion between the $00^0 1$ level and the lower $02^0 0$ and the $10^0 0$ levels in the CO_2 electronic ground state can be created. Laser oscillations can take place between the two multiple groups of transition near 9.6μ and 10.6μ.

In a mixture of CO_2-N_2 electron collisions with N_2 excite N_2 vibrational levels near $2330.7 cm^{-1}$. Then vibrational energy transfer from the excited N_2 to the $00^0 1$ vibrational levels in CO_2 can take place during collisions between N_2^* and CO_2. In an operating system laser action appears to be due to pumping through both direct electron pumping of the CO_2 and pumping via the N_2 route. Higher power and efficiency can be obtained in the CO_2-N_2 system due to more efficient pumping characteristics of the N_2 route.

It has been found that the addition of other gases can have beneficial effects. In particular the addition of He preferentially increases the rate of depopulation of the lower laser state, thereby having a direct effect on the operation of the laser. In addition, depending on the configuration and laser operating characteristics, the addition of He can also improve laser operation by modification of the velocity spectrum of the pumping electrons and by improving the conduction of heat from the excited gas to the walls. The relative proportions of gases and the total pressure of a mixture of CO_2:N_2:He affect the operation of the laser through complicated and detailed effects on the free electron energy distributions and the collisional couplings between the various gas molecules and ions in the system.[33-37]. These effects must be analyzed to determine the densities of the various excited molecules. The CO_2 molecules are, of course, also

coupled to the characteristics of the laser cavity through the effects absorption and stimulated emission of electromagnetic radiation present in the active region.

B. MECHANISM OF OPERATION OF THE CO LASER

As was previously mentioned, the operation of the typical CO laser involves an inversion of the population of individual rotational-vibrational levels involved in the laser action[38,41]. Figure 3 shows the energy level diagram for CO. Approximately 60 P branch line transition between rotational-vibrational levels up to the 17th vibrational level of the ground state have been observed to lase[42-47]. Due to the characteristic of the pumping mechanism transitions among low vibrational levels $v=1\sim5$ are difficult to observe.* The optical gain at the center of the $J\rightarrow J'$ rotational transition of the $v\rightarrow v'$ vibrational band is proportional to

$$G \propto N_v(2J+1)e^{-BJ(J+1)hc/kt} - N_{v'}(2J'+1)e^{-BJ'(J'+1)hc/kt} \qquad (7)$$

where N_v and $N_{v'}$ are the total population densities of the v and v' vibrational states, respectively, B is the rotational constant and equals about 1.92 cm^{-1}, and T is the rotational temperature (approximately equal to the gas collisional temperature). Due to the functional dependence on J in the exponent of Eq. 7, positive gains can be accomplished even for population densities where the ratio $N_v/N_{v'}$ is less than one. Due to resonant relaxation thermal equilibrium is rapidly achieved[50] in the CO vibrational system. Although lack of Boltzman type of equilibrium due to the anharmonic system should be accounted for in detailed analysis,[40,41,48] an estimate of the vibrational temperature required for positive gain can be obtained by using a Boltzman distribution for N_v and $N_{v'}$ in Eq. 7. For positive gain

$$T_v > \frac{1}{\frac{2B(J+1)}{GT_r} + \frac{K}{hcG}\ln\frac{2J+1}{2J+3}} \approx \frac{GT}{2B(J+1)} \qquad (8)$$

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*Note added in editing--These transitions have now been observed by N. Pjeu in a low temperature N_2-CO_2 transfer laser.

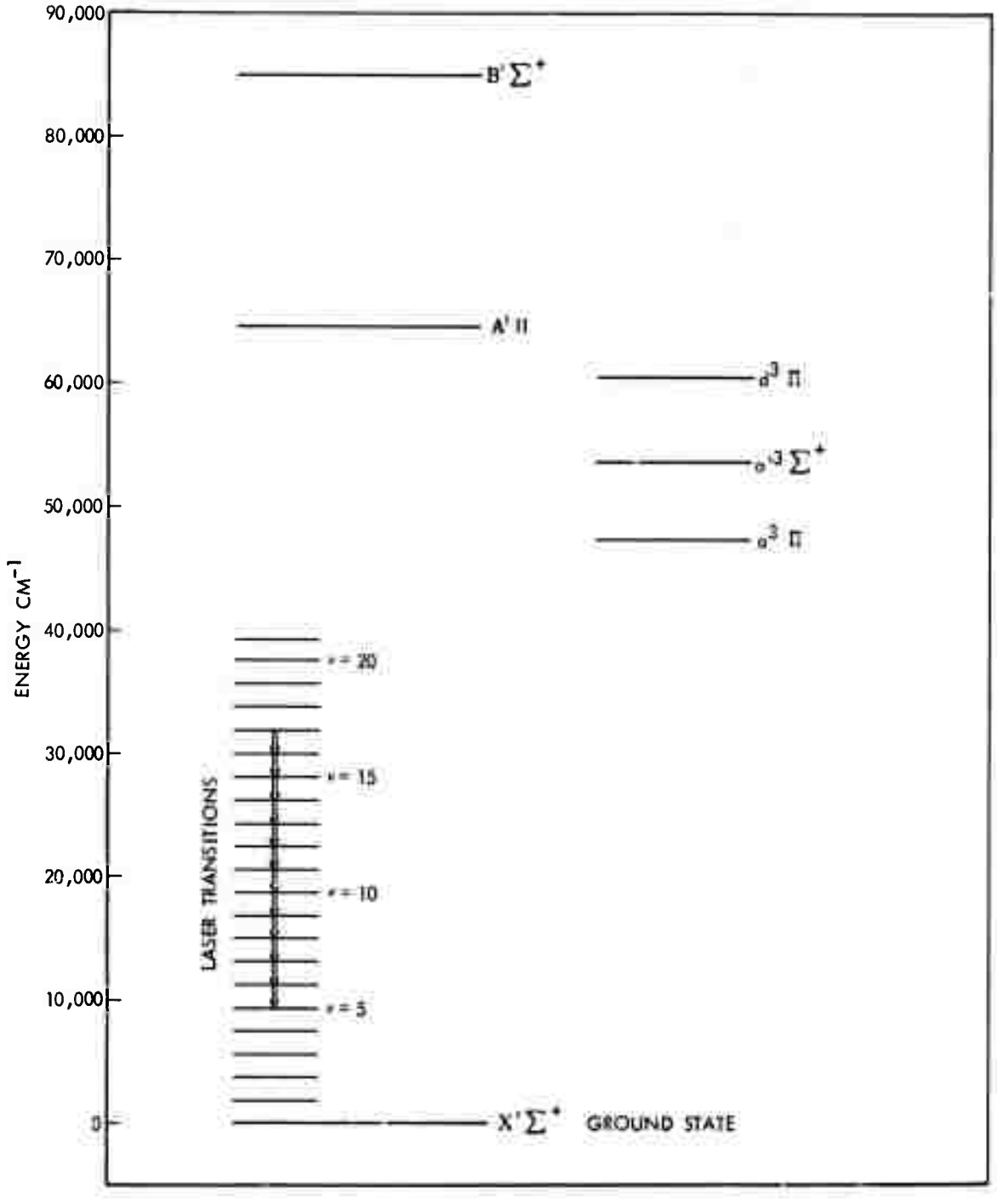

FIGURE 3. Energy States of the CO Gas Molecule Important in Gas Laser Operation. (Vibrational states are shown for the ground state only. The rotational states and many higher levels of the molecules are not shown.)

where G is the vibrational constant ($\approx 2143\ cm^{-1}$ for CO), the achievement of high values for T_v is difficult and, in fact, limited by the improved coupling to the translational motion in higher vibrational levels[49]. The requirement on the magnitude of T_r ($\simeq T$) can be greatly reduced by lowering the gas collision temperature. Several cascade chains have been observed in the operation of CO lasers. An example of a cascade is

$$[v=10,J=8] \xrightarrow[P10\text{-}9^{(9)}]{} [v=9,J=9] \xrightarrow[P9\text{-}8^{(10)}]{} [v=8,\ J=10] \xrightarrow[P8\text{-}7^{(11)}]{} [v=7,J=11]$$

$$\xrightarrow[P7\text{-}6^{(12)}]{} [v=6,\ J=12] \xrightarrow[P6\text{-}5^{(13)}]{} [v=5,\ J=13]. \qquad (9)$$

Time-dependence studies of pulsed laser output show the progress of the cascade clearly. In the above example the $P6\text{-}5^{(13)}$ transition lases and depopulates the $v=6$, $J=12$ level thereby increasing the gain for the $P7\text{-}6^{(12)}$ transition until laser action is possible. This process continues up the chain as a cascade from one vibrational level to the next producing laser action in sequence until the population density of the higher vibrational level is too low to produce sufficient gain for lasing.

Laser action by excitation of CO through electron collision has been accomplished in a glow discharge laser; however, measurements[38] strongly indicate that no direct excitation of CO molecules from ground state $v=o$ level can be the predominent pumping mechanism. Early measurement by Patel indicated that the electronic excitation of the $B^1\Sigma^+$ level with subsequent radiation to A^1II followed by radiative decay to various $X^1\Sigma^+$ vibrational levels constitutes the main excitation process in a pure CO laser. However, the addition of N_2 to CO allows for excitation of the CO vibration levels by vibrational energy transfer from nitrogen to CO according to the equations

$$N_2(\nu) + CO(\nu') \rightleftarrows N_2(\nu-1) + CO(\nu'+1) \quad (10)$$

$$N_2(\nu) + N_2(\nu') \rightleftarrows N_2(\nu-1) + N_2(\nu'+1) \quad (11)$$

$$CO(\nu) + CO(\nu') \rightleftarrows CO_2\ (\nu-1) + CO(\nu'+1) \quad (12)$$

Reactions 11 and 12 are rapid and result rapid relaxation to a Treamor-type thermal equilibrium distribution[40]. Reaction 10 is nearly resonant, the ν=1 N_2 level being 188 cm^{-1} higher than the ν^1=1 CO level. The relaxation constant of the reaction is approximately 1370 $torr^{-1}$ sec^{-1}. The addition of N_2 while resulting in improved CO laser operation complicates the situation by allowing the possibility of creation of atomic nitrogen, oxygen and the CN radical. These impurities can have important effects on the operation of the CO laser. CN in particular has a quenching action on the formation of population inversion buildup for three reasons: (1) its formation reaction is exothermic only if CO is vibrationaly excited to at least ν-13; therefore, its formation depopulates the high vibrational levels of CO; (2) CN in its ground electronic state can be excited to higher electronic states by vibrationaly excited N_2. These excited electronic states rapidly radiate and form a route for rapid depopulation of vibrational N_2 energy; (3) depending on geometric CN is believed to form deactivating deposits on the tube walls which progressively quench the laser effect.

The addition of other gas such as O_2, NO, Xe, He to the lasing mixture have experimentally been found to have beneficial effects and laser output can be experimentally optimized[50]. The addition of Xe is particularly interesting in that authors[51] report efficiency of 40 percent. These authors believe that the primary effect of Xe is to adjust the average electron energy in the flow discharge to bring the peak of the electron distribution function to closer coincidence with the peak of the electron excitation cross section for N_2 and CO.

IV. DESIGN CONSIDERATION FOR HIGH-POWER, ELECTRICALLY EXCITED GAS LASERS

In the previous section we discussed various developments which lead to the possibility of very high-power, electrically pumped gas lasers. At the present stage of development, the best method for obtaining such a device with a good optical quality beam and reasonable efficiency appears to consist of a pumping technique which uses ionizing radiation (probably high-energy electrons) to initiate and maintain an electron plasma in the active region of the laser. Figure 1 schematically illustrates typical arrangements. An electric field gradient applied to the active region accelerates the electrons of the plasma which, in turn, collide with gas molecules to provide the necessary molecular excitation for lasing. This technique allows the possibility of uniformly pumping reasonably large volumes of gas at high pressure. At present CO_2:N_2:He mixtures seem to provide the highest powers. However, it is not clear that future study on other gas mixtures cannot provide higher powers and higher efficiencies at different wavelengths. In order to achieve a high-repetition rate or continuous operation it seems likely that a system for flowing the gas through the active region will be required to provide cooling and perhaps purification of the gas. In the following sections we briefly discuss the design consideration and requirement on each of the subsystems of such a device.

A. POTENTIAL POWER AND ENERGY OUTPUT

From the point of view of determining the limits of power output the molecular gas laser can be considered in three types of operation: Q-Switched, Pulsed Quasi-Equilibrium, and Continuous Operation.

1. Q-Switched Operation

In the Q-Switched mode the excitation energy is supplied and stored in the gas until the Q of the cavity is switched to initiate lasing[52,53]. This type of operation provides the highest peak powers, particularly if a mode-locking system is included to achieve output pulses of short duration. Hole burning will limit the energy available in short pulses. The efficiency of converting electrical power to light energy is sacrificed. The amount of energy which can be stored depends on the pumping rate, the loss rate, and the number of lasing molecules in the active medium. Once the cavity is Q-Switched, lasing continues until the inversion is insufficient to provide the gain necessary for lasing. For example, we consider a system of N_2:CO_2 in the ratio 1:1. Although the use of pure CO_2 is possible the use of N_2 seems desirable to attain more efficient pumping. We assume pumping pulses which have a duration of a few times the time constant for deactivation of the v=1 vibrational states by collision. The energy transfer from vibrationally excited N_2 to vibrationally excited CO_2 is sufficiently rapid ($\sim$0.07μs·Atm) that these systems can be assumed to be near equilibrium at the end of the pumping pulse. Also decay of the lower laser levels in CO_2, which will be excited by electron collision, is sufficiently rapid that these levels can be assumed to be essentially unpopulated for low gas temperatures. In a simple two level system the ratio of excited molecule density to ground state molecule density is given by

$$\frac{N^*}{N_o} = \frac{R_s}{R_s+R_c}\left\{1 - e^{-(R_s+R_c)t}\right\} \qquad (13)$$

where R_s is the pumping rate and R_c is the decay rate. For the excited level to attain significant storage levels R_s should be comparable to R_c. In a high pressure CO_2 laser, the collision loss dominates other loss mechanisms. Moore et al.[33] measure the collision decay rate for CO_2 to be 2.68×10^5 sec^{-1} Atm^{-1}, and, therefore R_c is approximately 2.68×10^5 times the gas pressure. The effective pumping rate can be

determined from the electron density and average electron energy as described by various authors[54-56].

$$R_s \sim \left(\frac{\nu_{eff}}{N_j}\right) N_e \sim 10^{-8}\ N_e \qquad (13a)$$

where N_e is the electron density. If we assume we need $R_s \sim R_c$ this gives the required electron density. Under the above assumption the energy density available for laser output is approximately given by $\frac{1}{4}N_{CO_2}\ h\nu$ where N_{CO_2} is the density of CO_2 molecules and $h\nu$ is the quantum energy. Table 2 gives the maximum potential output energy and the required energy densities for CO_2:N_2 based on these approximations. The outputs listed in Table 2 are considerably higher than can be achieved in a practical laser.

TABLE 2. PARAMETERS FOR Q-SWITCHED CO_2:N_2 LASER OPERATION

Pressure (Atm)	Output Energy j/cm^3	Electron Density electron/cm^3	Current amp/cm^2	Gradient volt/cm
0.1	0.013	2.68×10^{12}	2.5×10	1.6×10^{4}
1.0	0.13	2.68×10^{13}	2.5×10^{2}	1.6×10^{5}
10.0	1.3	2.68×10^{14}	2.5×10^{3}	1.6×10^{6}

Also shown in the table is the current density required to attain the electron density specified assuming an average electron energy of one electron volt.

$$J = N_e\ \bar{u}\ e \qquad (14)$$

The voltage gradient which gives these conditions is tabulated in the last column of the table.

2. Pulsed Quasi-Equilbrium Operation

In this type of operation the Q of the cavity is not switched but is maintained at some appropriate value. Excitation energy is supplied to the cavity and once threshold is achieved laser action commences and continues until the excitation stops or conditions within the laser modify the threshold condition to the point where laser emission terminates. The rate equations governing this action for a CO_2:N_2:He laser form a complicated set of nonlinear integro-differential equations which have been studied by various authors (e.g., Cool[17], Moore[33], Rigrod[2,3]). Rate equations which include the dominant terms are given below. A more complete set of equations could be written which include the numerous possible states of excitation and ionization of the various molecules and the related excitation and relaxation processes. However, the detailed information on relaxation and excitation constants is not available for such a complete analysis. The symbols in the equations below are defined as follows: The collision relaxation constant for transfer of energy from A to B in a collision with C is devoted $K_{A\rightarrow B:C}$; thus $K_{N_2^2\rightarrow CO_2^3:CO_2}$ represents excitation of CO_2 to level 3 (see Fig. 2b) by a collision of an excited N_2^2 molecule with a CO_2 molecule. The concentration density (molecules/cm^3) of CO_2 or N_2 molecules in the ith level is denoted by (CO_2^i) and (N_2^i), respectively. The equilibrium value for the concentration density is denoted by a bar e.g., $\overline{CO_2^2}$; these are related to the total molecular densities by Boltzmann or Treanor distributions with appropriate temperatures. The coefficients for stimulated emission and absorption are S_{21} and S_{12} and the fraction of molecules in the levels 1 and 2 which are in the rotational states involved in lasing and are denoted by f_1 and f_2. The laser intensity in the cavity is denoted by I and the rate of excitation of nitrogen is $R_s(N_2^0)$ where R_s is approximately given by Eq. 13a. The rate equation for (CO_2^2) is then given by

$$\frac{d(CO_2^2)}{dt} = K_{N_2^1-CO_2^2:CO_2^0}\,(CO_2^0)\,(N_2^1) - K_{CO_2^2-N_2^1:N_2^0}\,(N_2^0)\,(CO_2^2)$$

$$- [K_{CO_2^2-CO_2^1:CO_2}\,(CO_2^0) + K_{CO_2^2-CO_2^1:He}\,(He) + K_{CO_2^2-CO_2^1:N_2^0}\,(N_2^0)]\,[(CO_2^2) - \overline{(CO_2^2)}]$$

$$- S_{21}\,If_2\,(CO_2^2) + S_{12}\,If_1\,(CO_2^1). \qquad (15)$$

In writing the equation, spontaneous radiative loss and gain and excitation from electron excitation of CO_2 have been neglected. The rate equation for (N_2^1) is

$$\frac{d(N_2^1)}{dt} = -K_{N_2^1-CO_2^2:CO_2^0}\,(CO_2^0)\,(N_2^1) + K_{CO_2^2-N_2^1:N_2^0}\,(N_2^0)\,(CO_2^2)$$

$$- K_{N_2^1-CO_2^1:CO_2^0}\,(CO_2^0)\,(N_2^1) + R_s(N_2^0). \qquad (16)$$

This neglects losses from collision of nitrogen molecules with nitrogen and helium molecules. These rates are known to be small. The rate for collision excitation of CO_2 into level 1 by collision with excited nitrogen could be large due to near-resonant nature of the reaction. Finally, the rate equation for (CO_2^1) is given by

$$\frac{d(CO_2^1)}{dt} = [K_{CO_2^2-CO_2^1:CO_2^0}\,(CO_2^0) + K_{CO_2^2-CO_2^1:He}\,(He) + K_{CO_2^2-CO_2^1:N_2^0}\,(N_2^0)]\,[CO_2^2 - \overline{(CO_2^2)}]$$

$$- [K_{CO_2^1-CO_2^0:CO_2^0}\,(CO_2^0) + K_{CO_2^1-CO_2^0:N_2^0}\,(N_2^0) + K_{CO_2^1-CO_2^0:He}\,(He)]\,[(CO_2) - \overline{(CO_2^1)}]$$

$$+ K_{N_2^1-CO_2^1:CO_2^0}\,(CO_2^0)\,(N_2^1) + S_{21}\,If_2\,(CO_2^2) - S_{12}\,If_1\,(CO_2^1). \qquad (17)$$

The sums $(CO_2^0) + (CO_2^1) + (CO_2^2)$ and $(N_2^0) + (N_2^1)$ are the total (CO_2) and (N_2) densities. To solve the equations for an oscillator arrangement an additional relation giving the interaction between the gain $S_{21}\ f_2\ (CO_2^2) - S_{12}\ f_1\ (CO_2^1)$ and the cavity parameters is required. For the oscillator case, assuming a uniform gain over a cavity of length L with effective reflectances $r_1\ r_2$, we have

$$S_{21}\ f_2(CO_2^2) - S_{12}\ f_1(CO_2^1) = \ln(r_1 r_2)/_2 h\nu L \qquad (18)$$

as the condition for steady oscillation.

In equilibrium we see that the population inversion is provided by collision pumping of the upper state [the first two terms on the right-hand side of Eq. 15] and by collision relaxation of the lower state [the second term in the brackets in Eq. 11]. Dissipative effects of collisions of excited CO_2 and excited N_2 molecules with various element of the gas mixture act to reduce the population inversion [the first term in brackets of Eq. 15 and the third term in Eq. 16].

In quasi-equilibrium operation, the output of the laser continues until the increase of the gas temperature due to conversion of energy to heat in the pumping and operating process causes the collision losses and the equilibrium density of the lower CO_2 state to increase to the point where lasing is turned off[5,57-60]. To a first approximation the collision constants K have a temperature dependence given by

$$K \simeq K_o \exp - (B/T^{1/3}) \qquad (19)$$

In Table 3 we give values for the coefficients K_o and B. Values in the tables were determined as follows. For $CO_2^2 \rightarrow CO_2^1 : CO_2^0$ we normalized the temperature dependence given by Burnett[61,62] to the relaxation value given by Moore[33]. Data for the temperature dependence of

of the various relaxation times is very limited[63,64]. For a crude approximation we assume B in Eq. 19 is proportional to the square root of the atomic weight of the molecule CO_2 is colliding with and use the relaxation times given by Moore; we also assume

$$K_{N_2^1 \rightarrow CO_2^1 : CO_2} \approx K_{CO_2^2 \rightarrow CO_2^1 : N_2}. \tag{20}$$

TABLE 3. RELAXATION COEFFICIENT FOR CO_2 RELAXATION

Reaction	K_o (Atm-μs)$^{-1}$	B (^oK)$^{1/3}$
$CO_2^2 \rightarrow CO_2^1 : CO_2$	64	36.6
$CO_2^2 \rightarrow CO_2^1 : N_2$	6.2	29
$CO_2^2 \rightarrow CO_2^1 : He$	0.33	11
$N_2^1 \rightarrow CO_2^1 . CO_2$	64	36.6

The rotational levels are assumed to be in equilibrium at the collision temperature T of the gas. Therefore, the value and temperature dependence of S_{12} and S_{21} are given by[17]

$$S_{21} = \frac{2.8 \times 10^5 \exp(-420/T)}{T} \tag{21}$$

and

$$S_{12} = 0.935\, S_{21}. \tag{22}$$

Finally, the temperature of the system is given by

$$T = T_o + \int_0^t \{R_s(N_2)\, Lh\nu_n - I\,(1-r)\}\, d\tau/CL \tag{23}$$

where C is the specific heat of the gas. The first and second terms in the brackets of Eq. 22, give the input and output power density, respectively.

The above set of equations (Eq. 15 through Eq. 23) have been numerically integrated to evaluate the properties and output from carbon-dioxide lasers with a variety of gas partial pressures, excitation rates, cavity lengths, and reflectivities. The results were analyzed to determine an optimum power density output configuration. Under equilibrium conditions the intensity from a laser oscillator is given by

$$I = I_s g_o L - I_s (1-R)/2 \qquad (24)$$

where g_o is the small signal gain and I_s is the saturation intensity at which the gain is reduced by a factor of two. In quasi-equilibrium operation I_s and g_o are functions of time through their dependence on the gas temperature. The integral of $I_s g_o$ is approximately the optimum energy density output W^{opt} which can be obtained from the laser gas.

Therefore,

$$W^{opt} = \int_{pulse} I_s g_o dt. \qquad (25)$$

In the calculations the partial pressures of helium, nitrogen, and carbon dioxide were optimized. In Table 4, we give the values for W^{opt} which were obtained for various rates of excitation and total gas pressures.

To minimize loss due to collision relaxation the intensity I must be maintained at a high level. The limits of energy output per pulse is determined by the limit placed on I and/or the pumping rate which can be accomplished without destruction of equipment. In practice, where good beam characteristics are required, practical limits

will be placed on the intensity I and the pumping rate by the requirements of uniformity of the active medium.

TABLE 4. OPTIMUM OUTPUT POWER DENSITY IN QUASI-EQUILIBRIUM OPERATION

Relative Partial Pressure $He:N_2:CO_2$	Total Pressure Atms	Electron Density 10^8 e/cm^3	w^{opt} joules/cm^3
1:1:1	1.00	10.0	1.81
	10.0	100.	18.5
1:2:3	1.00	10.0	1.76
	10.0	100.	8.45
2:3:1	1.00	10.0	2.10
	10.0	100.	14.4
3:1:2	1.00	10.0	1.49
	10.0	100.	11.6
1:3:2	1.0	10.0	2.04
	10.0	100.	21.7
3:2:1	1.00	10.0	1.86
	10.0	100.	11.5
2:1:3	1.00	10.0	1.46
	10.0	100.	7.01
1:1:2	1.00	10.0	1.61
	10.0	100.	10.0
1:2:1	1.00	10.0	2.08
	10.0	100.	19.1
2:1:1	1.00	10.0	1.69
	10.0	100.	12.7
1:1:3	1.00	10.0	1.50
	10.0	100.	8.43
1:3:1	1.00	10.0	2.23
	10.0	100.	16.7
3:1:1	1.00	10.0	1.61
	10.0	100.	9.32

3. Continuous Operation

In this type of operation the power level is maintained at a level low enough to permit an equilibrium gas temperature which allows continuous operation. This equilibrium temperature is determined by the level of pump power input and power loss by lasing, conduction or material flow. Rate equations which apply have already been discussed in the previous paragraphs.

B. REQUIREMENTS FOR A GOOD ELECTRON BEAM LASER PUMPING ARRANGEMENT

Many problems are encountered when one attemps to achieve high average power or high energy density in gas lasers. Requirements for good design include: (1) A good mechanical design is required to ensure a reliable and stable arrangement. (2) The design of the pumping configuration should provide for a uniform distribution of excited molecules in the active medium and should attain the highest possible efficiency. (3) For high average power devices generally a cooling system must be provided. (4) In gas lasers where a gas flow system is used, gas flow and density uniformity are required to achieve uniform pumping and uniform optical properties. (5) The design of the device itself and its ancillary equipment should be suitable for the purpose and use of the device and should be as simple to construct, assemble and repair as possible.

In the design of the pumping system of an electron beam laser which is to have a high-power output beam with good optical properties, utmost care must be taken to obtain uniform pumping with minimum thermal gradients in the gas. The electron economics and design consideration in the electron beam sustained discharge can be divided into two general areas: (1) High energy electrons which are defined as those electrons with sufficient energy to be easily capable of production of ionization and (2) Low energy electrons which are those that produce the gas excitation. This split of the electron distribution into two energy regions is, of course, possible only when two peaks which are fairly well isolated occur in the velocity distribution of the electrons.

1. Discussion of High Energy Electron Requirements

Table 5 lists mechanisms important in the determination of the spatial and velocity distribution of these electrons. Analytically $N(\bar{r},\bar{v})$, the velocity and spatial distribution of these electrons, can be determined by solving the transport equation for the boundary conditions and geometry of the laser. These equations should include terms which describe the various production, loss, and scattering processes[65-73] listed in Table 5. Fortunately, certain simplifying approximations can be made.

Non-uniformities in ionization resulting from non-uniformity in the spatial and velocity distribution of the high energy electrons have deleterious effects on the optical quality of the active medium. These effects are the result of change in the effective index of refraction as a result from thermal gradient and from changes in the composition of the gas.

The ionization per unit volume is given by

$$\text{Ionization/unit volume} = \int_{\text{velocity}} \Sigma\, N_i \sigma_i(v)\, n(\bar{r},\bar{v})\, v\, d\bar{r} \qquad (26)$$

where N_i is the density of gas molecules of excitation i and σ_i is their cross section for ionization. In the case of a composite gas a sum over the constituents is necessary. From the equation we see that variation in ionization density can result from spatial non-uniformity in the gas density N and the electron distribution function n. The cross section $\sigma_i(v)$ is a function of velocity. Therefore, changes in the velocity distribution with position also results in variation of the ionization density with position.

TABLE 5. FACTORS INVOLVED IN DETERMINING THE SPATIAL AND VELOCITY DISTRIBUTION OF HIGH ENERGY ELECTRONS

A.	Production	1) High energy electrons are injected into the area of the active medium after being externally produced by an external cathode and acceleration potential. The electron optics of this system determine the density and velocity distribution of the high energy electrons as they enter the laser active medium.
		2) Collisions between electrons in the gas molecules and the incident electrons can result in large energy transfers (up to 50% for elastic collisions). In these cases both the incident and the target electron can have sufficient energy to produce ionization; therefore, such collisions are a production mechanism for high energy electrons.
		3) If the field gradient in the active medium is sufficiently high the high energy tail of the low energy electron velocity distribution can contain electrons of sufficient energy to produce ionization. In a standard glow discharge laser this is the source of all ionizing electrons. In the electron beam sustained arrangement with high pressure gas any significant contribution of ionizing electrons from this source will result in gas breakdown and arcing.
		4) Photo production of electrons from internally produced photons (see for example D-3 below).
B.	Particle Loss	1) Loss through walls. In an ideal arrangement the high energy electrons all pass through the active medium and into the anode wall. Under practical situations the electrons are scattered and pass out of the active area through all its surfaces, depending on the transport equations. Scattering of electrons back into the active region also takes place at the walls and must be accounted for in the boundary condition.
		2) Loss due to decrease in energy below that required to produce ionization.
C.	Energy Gain	1) The field gradient in the active medium can make a significant contribution to the energy of the high energy electrons. Under certain conditions the energy lost due to the energy loss mechanisms can be compensated for by this gain. The energy gain is proportional to $\sin\theta$ where θ is the direction of the electron with respect to the field gradient.
D.	Energy Loss and Scattering	1) Inelastic electron scattering in which an incident electron scatters from an electron in a gas molecule causing ionization or molecular excitation is the predominant energy loss mechanism[75].
		2) Elastic scattering from electrons in the gas. In these hard collisions relatively large energy transfers and angular changes are made.
		3) Inelastic scattering from nuclei in the gas. For electron energies considered here this effect is smaller than either of the above. It is the source of bremsstrahlung radiation.
		4) Elastic scattering from nuclei in the gas. Depending on the angles of scattering considered this reaction is comparable to that in D-2 above. It causes large changes in angle but little change in energy.

The injection of high energy electrons from a specified location, and also the general geometric asymmetry of typical electron beam sustained lasers, generally make it impossible to achieve a truly uniform ionization density. Careful analysis is required to reduce the nonuniformities to a tolerable minimum. It should also be noted that $\sigma_i(v)$ depends on the excitation state of molecules in the gas. The ionization is coupled to the laser intensity I present in the cavity through this effect.

2. Discussion of Low Energy Electron Requirements

Table 6 lists mechanisms important in the determination of the spatial and velocity distribution of the low energy electrons. The velocity and spatial distribution of electrons can analytically be found by solution of the transport equation which they satisfy. Collision cross sections which are large and play the dominent role in determining the velocity distributions have been measured and calculated[54,55,67,68,76,77]. Nighan[54,55] has obtained approximate equilibrium distributions for the transport equation for the case of a uniform, steady electric field and has determined excitation efficiences for power transfer to excited CO_2 and N_2 vibrational states.

In reviewing the sources of pumping nonuniformities from low energy electrons we note that the rate of production of vibrationally excited molecules is the product of the density of unexcited molecules, the total electron density, and an effective collision frequency. Any local variation of these three parameters will result in a nonuniform excitation. Local variations of the total electron density can result from nonuniform ionization rate which has previously been discussed. The effective collision frequency depends on the velocity distribution of the electrons and is affected by local variation in the electric field gradient. Both of these parameters are modified by boundary and wall effects, by transport or diffusion phenomena, and by effects resulting from positive ion and electron space charges near boundaries. These parameters undergo large variations in the region of the cathode and to some extent the anode. Details of the

TABLE 6. FACTORS INVOLVED IN DETERMINING THE SPATIAL AND VELOCITY DISTRIBUTION OF LOW ENERGY ELECTRONS

A. Production

1) Ionization by high energy electrons

2) Ionization by the high energy tail of the low energy electrons.

3) High energy electrons which have been degraded by collision losses to the low energy category.

4) Wall effects - scattering of low energy electrons from the walls, production of low energy electrons by high energy electrons striking walls, production of low energy electrons by positive ions striking walls.

B. Loss

1) Collision with walls (including anode)

2) Recombination with positive ions. Various mechanisms for reaction are possible: a) Radiative recombination $x^+ + e \rightarrow x^* + h\nu$ b) dielectronic recombination $x^+ + e \rightarrow x_d \rightarrow x^* + h\nu$ where x^*_d is a doubly excited neutral state c) dissociative recombination $(xy)^+ + e \rightarrow y^* + x^*$ d) three body recombination $x^+ + e + z \rightarrow x^* + z$ or $x^+ + e + e \rightarrow x^* + e$ (McDaniel)[78].

3) Attachment to form negative ions. Various mechanisms for reaction are possible: a) radiative capture $e + A \rightarrow A^- + h\nu$ b) dissociative attachment $e + xy \rightarrow xy^{-*} \rightarrow x + y^-$ c) three body reactions $e + A + B \rightarrow B + A^-$ d) vibrational excitation and capture $e + xy \rightarrow [xy]^{-*}$; $[xy]^{-*} + A \rightarrow xy^- + A$ (McDaniel)[78].

C. Energy Gain and Loss

1) Field gradient action on electrons between collisions.

2) Elastic and inelastic electron-neutral, electron-electron, and electron-ion collision. Numerous reactions are possible in the energy range under consideration. Electron collision resulting in electronic and vibrational excitation dominate. Nighan[54,55] discusses the importance of the various reactions in determining the velocity distribution.

phenomena are similar to those which occur in the anode and cathode region of a glow discharge[79-81].

C. OUTPUT BEAM OPTICAL QUALITY

The optical quality of the output beam is determined by the amount of distortion which results from uncontrolled variations in the optical length across the wave front as it passes through the device[82,83]. The optical length is determined by the active medium, mirrors, window, nonactive gaps, and any other device placed in the path of the beam. Although careful attention should be given to all of these areas, we restrict our attention to the active medium region. The optical length through this region is determined by the physical length, L, and the refracture index, η. The phase shift, $\Delta\theta$, of the wave front from the ideal due to change in the optical path caused by index of refraction changes is given by[84]

$$\Delta\theta = \int_0^L \frac{2\pi s \Delta n ds}{\lambda} = \int_0^L \frac{2\pi s(n-1)}{\lambda} \left\{ \frac{\Delta N}{N} + \frac{\Sigma \Delta x_j \alpha_j}{\Sigma x_j \alpha_j} \right\} ds + \int_0^L \frac{2\pi s \Delta(W_e \alpha_e)\, ds}{\lambda} \tag{27}$$

We assume that for the gas mixture

$$\eta = 1 + 2\pi N \Sigma_j x_j d_j + N_e \alpha_e \tag{28}$$

where N is the total number of molecules per unit volume and Nx_j and α_j are the number of molecules per unit volume and the polarizability, respectively, for molecules of type j the term $\eta_e \alpha_e$ is the contribution to the index of refraction due to the electron gas. The sum is over the various gas molecules and each of their excitation states. For the lasing wavelength, polarizability of molecules in their various excitation states can be calculated with dispersion theory.

The angular distortion introduced by the phase shift $\Delta\theta$ is approximately $2\pi\lambda\Delta\theta/D$ where D is a characteristic aperture, therefore,

the distortion introduced is small compared to diffraction providing $\Delta\theta \sim 0.1$. Substituting values into Eq. 27 yields an upper limit of about 10^{-3} for the allowable variation of the term in the brackets. On examining Eq. 27 we see that distortions can arise from variations (i) in the total gas density, (ii) in the relative molecular excitation densities or (iii) in the free electron density. Total gas density variations can be the result of flow turbulence resulting from flowing gas as a result of gas replacement for high-power operation or as a result of thermal gradients. Such thermal gradients can result from nonuniform heating as a result of nonuniform pumping discussed in the previous section. Also power extraction during lasing can significantly change the gas heating in high efficiency gas lasers; thus the beam structure will have a significant effect on the temperature distribution through the active medium. In short pulsed operation density variations due to thermal gradients become insignificant because the disturbances propagate at approximately the velocity of sound ($\sim 2.5 \times 10^4$ cm/sec) and the lasing action is complete before significant density variations can build up.

Variations of relative molecular excitation densities on the other hand are generally essentially instantaneous and should be given consideration in the high-power laser design. Fortunately, the effects seem to be relatively small. These variations arise from 1) nonuniform pumping 2) nonuniform distribution of intensity across the beam and 3) at high temperature the density of rotational and low energy vibrational states may change significantly resulting in an instantaneous temperature effect. Each of these effects depends on the detail design of the system; gas composition, cavity arrangement, pump arrangement etc. They will become important as energy densities and gas pressure are increased.

V. SUMMARY AND RECOMMENDATIONS

Electron-beam-ionized, gas lasers offer the potential of attaining efficient, high-power lasers which produce light beams with good optical properties. In the previous sections of this report various aspects of the engineering design and operation of this type of a device have been discussed. As indicated in Section I some success has already been achieved in the design and operation of this type of laser. Although present information emphasizes carbon dioxide mixed with helium and nitrogen as the most suitable gas for high-power application, further study of some of the likely candidates listed in Table 1, or some other as yet unstudied gas species, may develop another gas or gas mixture suitable for use in the production of high-power gas lasers.

In the development of the carbon dioxide system for high-power application considerably more study is required in the following areas: (a) the measurement of basic physical processes and development of analytical techniques, (b) the development and testing of materials and components for construction of this type of laser, and (c) the overall system engineering design and prototype construction and testing. Each of these will be briefly discussed in the following paragraphs.

As has been discussed in Sections III and IV information on numerous basic physical processes must be developed for the analysis and design of electron-beam-ionized, gas lasers. In particular, only fragmentary information exists on the absolute value and temperature dependences of the collision relaxation and excitation constants of the type discussed in Section III-A. This information is vital in the analysis of laser design and operation. Ideally the velocity dependence of the molecule-molecule cross sections for each of the numerous

processes and molecular species involved would be desirable. From these cross sections the various relaxation and excitation constants and their temperature dependence can be calculated. However, since measurement of these cross sections is difficult, and in some cases impossible, direct measurement of these rate constants versus temperature is the only way to obtain the required information. The dominant electron-molecule cross sections required for design of the excitation electron beam and the related electron plasma as discussed in Section IV-B have been measured over most of the regions of interest. Some effort should be expended checking and improving the accuracy and resolution of these measurements. Also measurement for each of the molecular species and their excited states, should proceed on some of the as yet unmeasured cross sections required to allow more detailed analysis of the various effects listed in Tables 5 and 6.

Engineering information and development are required on the materials and components used in construction of the carbon dioxide laser. The optimum design and resulting limits of the optical beam power and energy density should be determined for various optical components such as mirrors, windows, transmission media, etc. Cathode, anode and related electron beam components need to be developed for production of the excitation condition required for laser operation as discussed in Section IV-B. A study should be made of material problems which develop as a result of chemical reactions and shock in the environment of large temperature and pressure extremes, high optical radiation intensity, high molecular excitation or ionization densities, and high electron densities existing in the laser cavity and associated components. Also study should be made of special material requirements which result from gas flow, cooling, and purification criteria.

Before construction of large devices can be carried out, careful system design and development studies must be pursued. Parametric design studies should be completed on a number of different overall concepts for a large system. After investigating the engineering features, problems, and costs of each design a choice can be made of

the design which best suits the requirements. For further development prototype construction should proceed to: (1) test modules, components, and materials for use in a large system, (2) test and check the result of engineering calculations and predictions on the behavior of the device, and (3) measure basic physical quantities of interest for the design of a larger system.

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